

ROSEN  
Appl. No. 10/543,095  
November 26, 2008

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**AMENDMENTS TO THE CLAIMS:**

This listing of claims will replace all prior versions, and listings, of claims in the application:

42 (currently amended). A method for the preparation of a supported catalyst composition suitable for the oxidation of ethane to ethylene and/or acetic acid, and/or the oxidation of ethylene to acetic acid, said supported catalyst composition comprising a catalyst comprising one or more metal components, supported on a support comprising alpha-alumina, which method comprises:

(a) forming a slurry of the one or more metal components and alpha-alumina support particles or an alpha-alumina support precursor; and

(b) spray-drying the slurry;

wherein the catalyst is selected from a catalyst which comprises palladium as the metal component and a catalyst which comprises a combination of molybdenum, vanadium and niobium as the metal components.

43 (currently amended). The method according to claim 42, which further comprises:

(~~a~~ c) calcining the spray-dried slurry.

44 (currently amended). The method according to claim 42, wherein the alpha-alumina used for the support has a surface area, as measured by BET, of less than 15 m<sup>2</sup>/g, ~~such as less than 10 m<sup>2</sup>/g, for example, less than 5 m<sup>2</sup>/g.~~

ROSEN  
Appl. No. 10/543,095  
November 26, 2008

45 (currently amended). The method according claim 42, wherein the alpha-alumina used for the support has a surface area, as measured by BET, of at least 0.1 m<sup>2</sup>/g, ~~most preferably at least 0.5 m<sup>2</sup>/g, such as in the range 0.5 m<sup>2</sup>/g to less than 10 m<sup>2</sup>/g, more preferably in the range 0.5 m<sup>2</sup>/g to less than 5 m<sup>2</sup>/g.~~

46 (currently amended). The method according to claim 42, wherein the alpha-alumina used for the support has a density of between 0.5 and 5g/cc, ~~preferably between 0.8 and 2 g/cc.~~

47 (previously presented). The method according to claim 42, wherein the support is alpha-alumina.

48 (previously presented). The method according to claim 42, wherein the support comprises a mixture of alpha-alumina with one or more non-alpha-alumina materials.

49 (previously presented). The method according to claim 48, wherein the support comprises one or more alpha-aluminas in combination with one or more silicas and wherein the one or more silicas are low sodium-containing silicas.

50 (currently amended). The method according to claim 48, wherein alpha-alumina comprises at least 10% by weight of the total support, ~~preferably at least 20%,~~

ROSEN  
Appl. No. 10/543,095  
November 26, 2008

~~such as 40% or more, and most preferably 50% or more by weight of the total weight of the support.~~

51 (currently amended). The method according to claim 42, wherein the supported catalyst composition has a surface area, as measured by BET, of between 0.1 and 20 m<sup>2</sup>/g, ~~more preferably between 1 and 5 m<sup>2</sup>/g.~~

52 (currently amended). The method according to claim 42, wherein the supported catalyst composition has a density of between 0.5 and 5g/cc, ~~more preferably between 0.8 and 2 g/cc.~~

53 (currently amended). The method according to claim 42, wherein the one or more metal components are present in the supported catalyst composition in a total amount equivalent to between 5% and 60% by weight of the total supported catalyst composition, ~~preferably between 20 and 50% inclusive by weight.~~

54 (canceled).

55 (currently amended). The method according to claim ~~54~~ 42, wherein the catalyst is represented by the formula Mo<sub>a</sub>Pd<sub>b</sub>X<sub>c</sub>Y<sub>d</sub> wherein X represents one or several of Cr, Mn, Nb, Ta, Ti, V, Te and W; Y represents one or several of B, Al, Ga, In, Pt, Zn, Cd, Bi, Ce, Co, Rh, Ir, Cu, Ag, Au, Fe, Ru, Os, K, Rb, Cs, Mg, Ca, Sr, Ba, Nb, Zr, Hf, Ni, P, Pb, Sb, Si, Sn, Tl and U and a=1, b=0.0001 to 0.01, c = 0.4 to 1 and d = 0.005 to 1.

ROSEN  
Appl. No. 10/543,095  
November 26, 2008

56 (canceled).

57 (currently amended). The method according to claim 42, wherein the catalyst comprises the metals molybdenum, vanadium, niobium and gold in the absence of palladium according to the empirical formula:



wherein Y is one or more metals selected from the group consisting of: Cr, Mn, Ta, Ti, B, Al, Ga, In, Pt, Zn, Cd, Bi, Ce, Co, Rh, Ir, Cu, Ag, Fe, Ru, Os, K, Rb, Cs, Mg, Ca, Sr, Ba, Zr, Hf, Ni, P, Pb, Sb, Si, Sn, Tl, U, Re, Te and La;

a, b, c, d, e and f represent the gram atom ratios of the metals such that:

$$0 < a \leq 1; 0 \leq b < 1 \text{ and } a + b = 1;$$

$$10^{-5} < c \leq 0.02;$$

$$0 < d \leq 2;$$

$$0 < e \leq 1; \text{ and}$$

$$0 \leq f \leq 2.$$

58 (previously presented). The method according to claim 57, wherein  $a > 0.01$ ,  $d > 0.1$  and  $e > 0.01$ .

59 (previously presented). The method according to claim 58, wherein  $e \leq 0.5$  and  $0.01 \leq f \leq 0.5$ .

ROSEN

Appl. No. 10/543,095

November 26, 2008

60 (previously presented). The method according to claim 59, wherein  $0.4 \leq d \leq 0.865$ ;  $0.135 \leq e \leq 0.23$ ; and  $0.55 \leq d + e \leq 1$ :

61 (previously presented). The method according to claim 60, wherein  $a > 0.01$ ,  $0.0001 < c \leq 0.002$ ,  $0.425 \leq d \leq 0.8$ ,  $0.14 \leq e \leq 0.20$ ,  $0.6 \leq d + e \leq 0.95$ , and  $f \leq 0.2$ .

62 (previously presented). The method according claim 61, wherein  $0.0005 < c \leq 0.001$ ,  $0.45 \leq d \leq 0.7$ ,  $e \geq 0.15$ ,  $d + e \leq 0.9$ , and  $f \leq 0.02$ .

63 (previously presented). The method according to claim 62, wherein  $d \geq 0.5$ ,  $e \leq 0.18$ , and  $d + e \geq 0.7$ .

64 (previously presented). The method according to claim 63, wherein  $d + e \geq 0.8$ .

65 (previously presented). The method according to claim 57, wherein  $a = 1$ .

66 (previously presented). The method according to claim 57, wherein Y is selected from the group consisting of Sn, Sb, Cu, Pt, Ag, Fe and Re.

67 (previously presented). The method according to claim 42, wherein the step (a) comprises (i) preparing separate solutions comprising each metal compound by dissolving sufficient quantities of soluble compounds and/or dispersing any insoluble

ROSEN  
Appl. No. 10/543,095  
November 26, 2008

compounds or quantities of said compounds so as to provide a desired gram-atom ratio of the metal components in the catalyst composition, (ii) wherein the catalyst comprises more than one metal component, mixing the respective solutions to form a single solution comprising the desired quantities of metal components, and (iii) mixing the resulting solution with alpha-alumina support particles or alpha-alumina precursor, and if required, other support materials or precursors, to form a slurry.

68 (previously presented). The method according to claim 67, wherein the one or more solutions comprising the metal components are aqueous solutions having a pH in the range from 1 to 12, preferably from 2 to 8, at a temperature of from 20° to 100°C.

69 (currently amended). The method according to claim 42, wherein the spray-drying is performed at an outlet temperature of at least 100°C, ~~preferably between 120°C and 180°C, for example, between 130°C and 150°C.~~

70 (currently amended). The method according to claim 42, wherein the spray-drying is performed at an inlet temperature of between 250°C and 350°C, ~~for example, between 280°C and 300°C.~~

71 (withdrawn). A supported catalyst composition suitable for the oxidation of ethane to ethylene and/or acetic acid, and/or the oxidation of ethylene to acetic acid, characterised in that the supported catalyst composition has been prepared according to the method of claim 42.

ROSEN  
Appl. No. 10/543,095  
November 26, 2008

72 (withdrawn). A process for the selective oxidation of ethane to ethylene and/or acetic acid, and/or the selective oxidation of ethylene to acetic acid which oxidation process comprises contacting ethane and/or ethylene with a molecular oxygen-containing gas at elevated temperature in the presence of a spray-dried supported catalyst composition as claimed in claim 71.

73 (withdrawn). The process according to claim 72, wherein the supported catalyst composition has been calcined by heating at a temperature in the range from 250 to 500°C in the presence of an oxygen-containing gas, for example air.

74 (withdrawn). The process according to claim 72, which is a fluidised bed process.

75 (withdrawn). The process according to claim 74, wherein the particle size of the supported catalyst composition is such that at least 50% of the particles have a size less than 300 microns, and preferably such that at least 90% of the particles have a size of less than 300 microns.

76 (withdrawn). The process according to claim 75, wherein the supported catalyst composition is in the form of microspheroidal particles.

77 (previously presented). The process according to claim 72, wherein the

ROSEN  
Appl. No. 10/543,095  
November 26, 2008

molecular oxygen-containing gas is oxygen or oxygen diluted with a suitable diluent, such as nitrogen.

78 (withdrawn). The process according to claim 72, wherein there is fed, in addition to ethane and/or ethylene and the molecular oxygen-containing gas, water (steam).

79 (withdrawn). The process according to claim 72, wherein there is fed to the process a feed composition (in mol%) comprising 40 to 80% ethane, 0 to 10% ethylene, 0 to 20% water, 2 to 10% oxygen and with a balance of inert gas, preferably nitrogen.

80 (withdrawn). The process according to claim 72, wherein the elevated temperature is in the range from 200 to 500°C, preferably from 200 to 400°C, and most preferably in the range of 260°C to 360°C.

81 (withdrawn). The process according to claim 72, wherein the process is operated at a pressure in the range from 1 to 50 bar, preferably from 1 to 30 bar.

82 (withdrawn). The process according to claim 72, wherein the process is operated with a gas hourly space velocity (GHSV) of between 100 and 10,000 h<sup>-1</sup>, preferably 1000 to 5000 h<sup>-1</sup>.